Correlative, Static and Dynamic Properties of Near-Critical Liquids in Small Volumes of Rectangular and Round Section[1]

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ABSTRACT

In the given paper we report the declination of the critical temperature, density, viscosity and susceptibility of the one-component near-critical liquid in the small volumes of bar and cylinder geometry. Geometrical factor K which sets the degree of the spatial limitation defined by $K=q/Rco$, were the q is a characteristic size of a system. New values of critical parameters were defined by the maximum of the correlation length. Our results suggest that the growth behavior of correlation length should remain only along the axis associated with the present geometry, i.e. in spatially unlimited direction, moreover not at the critical temperature of bulk liquid, but at the new one. Reduced geometry of a system leads to decreasing of the critical parameters like the critical temperature and density while characteristic size of a system becomes smaller.

In contrast to a spatially infinite system for which the singular part of the viscosity demonstrates a "weak" anomalous growth at the bulk critical temperature, the maximum value of the viscosity in a finite-size liquid at the bulk criticality turns out to be finite and dependent on the characteristic size of a system as $\eta{\sim}q^{x}$, where x=0.06 is the universal exponent.

The conducted research let us to make the conclusion that reducing of the volume's size leads to decreasing of the susceptibility after the decreasing of the correlation length. However, its anomalous growth manifests at the new critical temperature which is the same one, as it was to be expected, which is calculated for the present geometrical conditions of spatial limitation of a system.

KEY WORDS: correlation length, critical temperature, cylinder, exponents, finite size, viscosity.

1. INTRODUCTION

Phase transitions and critical phenomena in finite-size systems have enough many specific particularities in contrast with similar phenomena in unlimited systems. Exactly for spatially limited systems exist an absence of the long-range nature of correlations between order parameter fluctuations along direction of spatial insufficiency, shift of critical singularities, interesting peculiarities of critical light opalescence. The direct consequences of system's spatial limitation are declination of the critical parameters (critical temperature T_c, density ρ_c, concentrations, etc.), critical exponents and some other important characteristics as, for example, viscosity of being under investigating small volume of liquid.

Anomalies, which are observe under the second-order phase transition and critical phenomena in space unlimited systems, greatly change its nature in those cases when a matter becomes limited in one or two dimensions. For systems, which are space limited in all three dimensions and which have distinctive single-line sizes smaller or comparable with maximal attainable value of correlation length, have to expect (and this is confirmed by many calculations) disappearances of critical anomalies.

Let us now use the results of correlation properties analyses for finite size cylindrical system [1] in order to examine the location shift of critical maximum of correlation length of density fluctuations. We will consider being near to the critical condition one-component liquid, which filled in cylindrical sample, which has radius a and extends infinitely along the axes, i.e. $0<x,y\leq a$, $-\infty<z<\infty$. So we have one infinite direction along the axis of the cylinder, which makes the condition of thermodynamic limit satisfied and we can investigate critical behavior such kind of system directly. This set of a problem let us to avoid inconveniences as with Monte Carlo simulation which is always in really finite system and have required extrapolation of obtained results to infinite volume [2] using widely accepted methods like finite-size scaling theory [3,4] and Binder's methods [5,6] in order to obtain critical characteristics.

2.1. SHIFT OF CRITICAL TEMPERATURE

For sufficiently close vicinity of critical isochore a new critical temperature $T_c(K)$, can be determined as under which must be observe anomalous growth of the correlation length component $(R_c)_z$ along the cylinder axis. Correlation length R_c in spatially limited system of cylindrical geometry appears dependent not only on thermodynamic variables (temperature, density and etc.), but also on the geometrical factor K, having the value of molecular layers number, which is possible to arrange along the cylinder radius. In contrast to spatially unlimited system in a cylindrical sample of radius *a* the longitudinal component of correlation length $(R_c)_z$ in bulk criticality have a finite value and is determined by expression [1]

$$(R_c)_z=R_{co}\cdot K\cdot(K^2\cdot\tau^{2\nu}+\psi_1^2)^{-1/2} \quad \text{for } \tau>0 \quad \text{and}$$

$$(R_c)_z=R_{co}\cdot K\cdot(\psi_1^2-K^2\cdot\tau^{2\nu})^{-1/2} \quad \text{for } \tau<0 \qquad (1)$$

where R_{co} is the amplitude of correlation length, $K=a/R_{co}$ - the geometrical factor, temperature variable $\tau=(T-T_c)/T_c$, ν is the critical exponent of correlation length. In general case of the boundary condition of first kind for pair correlation function $G_2(a,z)=F(z)$ the naught ψ_1 is the solution of following transcendent equation at $\tau>0$:

$$J_0(\psi_1)\cdot e^{-1}-F((R_{co}^{-2}\cdot\tau^{2\nu}+\psi_1^2\cdot a^{-2})^{-1/2})=0 \qquad (2)$$

where $J_0(u)$ - cylindrical first kind's Bessel function of zero order. The expression for function F(z) will depend on particular statements of a problem.

In accordance with criterion of maximum in the correlation length and using the Eq. (1) we have for the region $\tau<0$:

$$\psi_1^2 \cdot K^{-2} - \tau^{2\nu} = \tau^{*2\nu} \quad (3)$$

where $\tau^* = (T - T_c^*(K))/T_c^*(K)$ - the temperature variable for spatially limited system. It is necessary to notice that the used above defining condition (3) of critical temperature mean physically, that at the achievement of the new critical temperature $T_c^*(K)$ of a spatially limited liquid in the sample of cylindrical geometry might exist anomalous growth of the longitudinal component of correlation length (1) along the cylinder axis. From Eq. (3) we can get the following formula for the critical temperature of liquid in the small volume with cylindrical geometry:

$$T_c^*(K) = T_c \cdot [1 + (\psi_1/K)^{1/\nu}]^{-1} \quad (4)$$

From Eq. (4) naturally follows that under transferring to the spatially unlimited system when the radius of cylinder aspire to infinity ($a \to \infty$) accompanied by geometrical factor ($K \to \infty$), the new critical temperature $T_c^*(K)$ became equal to the bulk critical temperature T_c, i.e. the shift will be absent. Results of calculation of the new critical temperature are in a good agreement with [7,8] where size-dependent shift of the "effective critical temperature" were defined by the maximum in the specific heat from scaling arguments.

The analysis of Eq. (4) shows that the shift of critical temperature of cylindrical sample $T_c^*(K)$ from critical temperature T_c of volumetric phase may be highly considerable. For example, in the case of zero boundary condition under Tc=300 K , geometric factor K=100 and mean-field value of exponent $\nu = 0.5$ difference of critical temperature is $\Delta T_c = T_c - T_c^*(K) = 0.173°K$. It correspond to the shift of critical point on $\Delta\tau = 6.4 \cdot 10^{-5}$ lower then bulk location.

From standpoints of existing theories, in which was conduct evaluation of shift of critical temperature in spatially limited systems, Eq. (4) being in agreement with results of papers [9-11] and exactly for value

$$(T_c(\infty)-T_c(n))/T_c(\infty)=b\cdot n^{-\lambda} \tag{5}$$

where n - a number of atomic layers and b is coefficient. In papers of Binder and Hoenberg [9], Domb [10], Fisher and Barber [11] was shown that exponent $\lambda=1/\nu$. However, Eq. (5) does not agreed with initial results of work Fisher and Ferdinand [12], where $\lambda=1$.

It is necessary to notice that dependency of shift of critical temperature ΔT_c on L~n with the critical exponent $\Theta=1/\nu$ possibly to confirm with using of scaling hypotheses for finite-size systems [13]. Let assume that for the unlimited system ($V=L^d\rightarrow\infty$, d - space dimensionality) phase transition occurs at temperature $T=T_c(\infty)$. For limited system with characteristic size L phase transition turns out to be expanded, and it is described by the vicinity

$$\Delta T_{vic.}\sim L^{-\Theta T} \tag{6}$$

where Θ_T - certain critical index. The center of phase transition area, more exactly - corresponding temperature $T_c(L)$, turns out to be shifted on value

$$T_c(\infty)-T_c(L)\sim L^{-\lambda T} \tag{7}$$

where λ_T - one more critical index which defining particularities of critical behaviour of finite-size systems. Using sufficiently easy scaling considerations it is possible to find numerical values of critical indexes Θ_T , λ_T. All distinctive critical singularities are bound, as is well known, with the correlation length ξ of order parameter fluctuations, which for unlimited systems is described by the formula:

$$\xi=\tau^{-\nu}\cdot\xi_1(h/\tau^{\beta\delta})=h^{-\nu/\beta\delta}\cdot\xi_2(\tau/h^{1/\beta\delta}) \tag{8}$$

Here ν, β, δ - critical exponents, h - an external field is associated to the order parameter, but $\xi_1(x)$ and $\xi_2(y)$ - scale functions, having following asymptotics:

$$\xi_1(x\to 0)=R_{co} \qquad \xi_2(y\to 0)=R_{co}$$

$$\xi_1(x\to\infty)\sim x^{-\nu/\beta\delta} \qquad \xi_2(y\to 0)\sim y^{-\nu} \tag{9}$$

On this basis, as well as on the background of considerations of spatial dimensionality let us assume value of the characteristic size of a system approximately equal to the correlation length ξ ($L\approx\xi$). Then for the system, which is under the zero field h=0 (it is the reality for the liquid in vicinity of the critical isochore $|\rho-\rho_c|<<\tau^{\beta}$ or for magnetic in "weak" magnetic field $H<<\tau^{\beta}$), get $\xi=R_c\cdot\tau^{-\nu}$, $\tau\sim\xi^{-1/\nu}$, that gives us

$$\Delta T_{vic.}\sim\tau\sim\xi^{-1/\nu}, \qquad [T_c(\infty)-T_c(L)]\sim\tau\sim\xi^{-1/\nu} \tag{10}$$

Comparing Eq. (6),(7) with Eq. (10), respectively, get for sought critical indexes following results: $\Theta_T=1/\nu$, $\lambda_T=1/\nu$. Similar results could be received for phase transitions induced by the external field h (it is real - for liquids under $T=Tc(\infty)$ in gravitation field, for magnetics in "strong" magnetic field $H>>\tau^{\beta}$).

Also the important consequence from Eq. (4) is that fact, that it base in packed consensus with experimental data of Lutz with co-authors [14]. Really, shift of critical temperature are characterized by the dependency on the geometric factor K and, consequently, on the linear size L toward spatial insufficiency of a system. This dependence kept an inverse value of critical exponent ν. Other our result, which is confirm by the preceding theoretical and experimental works (references in [13]), is that that in the small volume in contrast with the critical temperature T_c of unlimited (volumetric) phases the shift of critical temperature $\Delta T_c(L)$ of liquid is in the direction of decreasing, i.e. $T_c(L)<T_c$.

We can bring else some additional theoretical considerations in favour of reducing of the critical temperature $T_c(L)$ of space-limited systems with respect to its value $T_c(\infty)$. Well known (refer for instance to [15]), that condition of achievement of critical temperature in mean-field approximation is a performing a following expression:

$$N\cdot\varphi_{cp}/k_B\cdot T_c=1 \qquad (11)$$

where N - number of connections (couple interactions) , and φ_{cp} - energy of couple interaction. Obviously, that as a result of spatial restriction of system (for instance, when system turning to the plane-parallel sample by the thickness L, which contains several monolayers) number of connections N became smaller effectively. Then from the condition (11) immediately follows a decreasing of critical temperature $T_c(L)$ for the matter in the small volume.

In the thermodynamic limit first-order phase transitions are characterized by δ-function singularities in the second derivatives of the free energy at the transition point. However, in finite-size systems δ-function singularities are rounded and the effective transition point is shifted. These behaviors at the first-order transitions in finite-size systems are qualitatively similar to the finite-size effects at second-order transitions. In [16] were presented results for the Ising model in an external magnetic field, which exhibits a first-order phase transition below the bulk critical temperature . In the theory of Fisher and Berker [17], finite size scaling at the first-order transition is treated identically to scaling at a second-order transition.

2.2. SHIFT OF CRITICAL DENSITY

Using the similar way as above for temperature we can conduct consideration of the changing of a new critical density $\rho_c^*(K)$ in spatially limited system of cylindrical geometry in contrast with the value of critical density ρ_c for unlimited volumetric phase. The only difference is connected with changing of temperature dependence of correlation length $R_c \sim \tau^{-\nu}$ on the corresponding density dependence. Employed scaling-low equation of state [18] $\Delta\rho = B_p \cdot (-\tau)^{\beta}$, where $\Delta\rho = (\rho - \rho_c)/\rho_c$ - deflections of density ρ of liquid from critical value ρ_c, B_p - is a proportionality constant and β is a critical exponent, we can write $R_c \sim \Delta\rho^{-\nu/\beta}$. Then in vicinity of a critical isotherm, where $\Delta\rho >> \tau^{\beta}$, for single-component liquid new value of critical density became equal:

$$\rho_c^*(K) = \rho_c \cdot [1 + (\psi_1/K)^{2/(\delta-1)}]^{-1} \qquad (12)$$

for the constant boundary condition. From Eq. (12) naturally follows that under transferring to the spatially unlimited system ($K \rightarrow \infty$), the $\rho_c^*(K) \rightarrow \rho_c$ and the shift of density is absent. The analysis of Eq. (12) shows that the shift of critical density of cylindrical sample $\rho_c^*(K)$ from critical density ρ_c volumetric phase can turn out to be highly significant. So, in the case of zero border condition under $\rho_c = 300$ kg·m^{-3} (such value is typical for some hydrocarbons), geometric factor K=1000 and mean-field values of exponents $\nu = 0.5$ and $\beta = 0.5$ $[2/(\delta-1) = \beta/\nu]$ the gap of critical density is $\rho_c - \rho_c^*(K) = 0.72$ kg·m^{-3}.

In [19] from the Monte Carlo calculations for fluid in a porous material is obtained that the vapor-liquid coexistence region appears at lower temperatures than for the bulk, also the condensed phase densities are lower than those in the bulk. There mentioned that primary conclusion from their work: the critical temperature is lower than that in the bulk. They noted that the simple mean-field theory can predict the suppression of the critical temperature and lowering of the critical density for such kind of systems.

2.4. CHANGE OF VISCOSITY

Using the results is presented above we will investigate the change of viscosity η due to the spatial insufficiency. By combining of scaling relation for viscosity in the absence of shear [20]

$$\eta=\eta^{B}\cdot(Q_0\cdot Rc)^{x} \qquad (13)$$

and Eq. (1) for the correlation length for the case of zero boundary condition we can get an equation for viscosity η^* of a spatially-limited liquid cylindrical system of radius *a* in the region $\tau<0$, i.e. in the near-critical state in a form

$$\eta^*=\eta^{B}\cdot A\cdot K^{x}\,(\mu_1^{2}-K^{2}\cdot\tau^{2\nu})^{-x/2} \qquad (14)$$

where η^B is a background viscosity, x=0.06 - critical exponent, μ_1=2.4048 is first naught of J_0 Bessel function and $A=(Q_0\cdot Rc)^x$ - system-dependent constant. As it possible to see from Eq. (14), the viscosity in spatially limited system of cylindrical geometry depends not only on thermodynamic variable, but also on the geometric factor K. In contrast from spatially unlimited system, for which viscosity increase under the critical temperature ($T\rightarrow T_c$, $\tau\rightarrow0$) up to infinity in conformity with the formula [20,21] $\eta/\eta^B\sim\tau^{-\phi}$ (ϕ=0.41 – is an exponent), in the cylindrical sample of radius *a* maximum value of viscosity for $\tau\rightarrow0$ appear finite and equal to

$$\eta^*(\tau=0)\sim0.95\cdot a^{x}\cdot\eta^{B}\cdot Q^{x} \qquad (15)$$

Limiting transition to the case of spatially unlimited system at $K\rightarrow\infty$ have to be realized. From the Eq. (14) it is visible, that this transition takes place, i.e. $\eta^*\sim\tau^{-x}$ for $K\rightarrow\infty$. It is possible to make a conclusion that viscosity became smaller in course of reducing of size of the system following the reducing of correlation length. The Fig. 1 illustrates temperature dependence of η^* at K=300 and ν=0.5 for the cylindrical sample

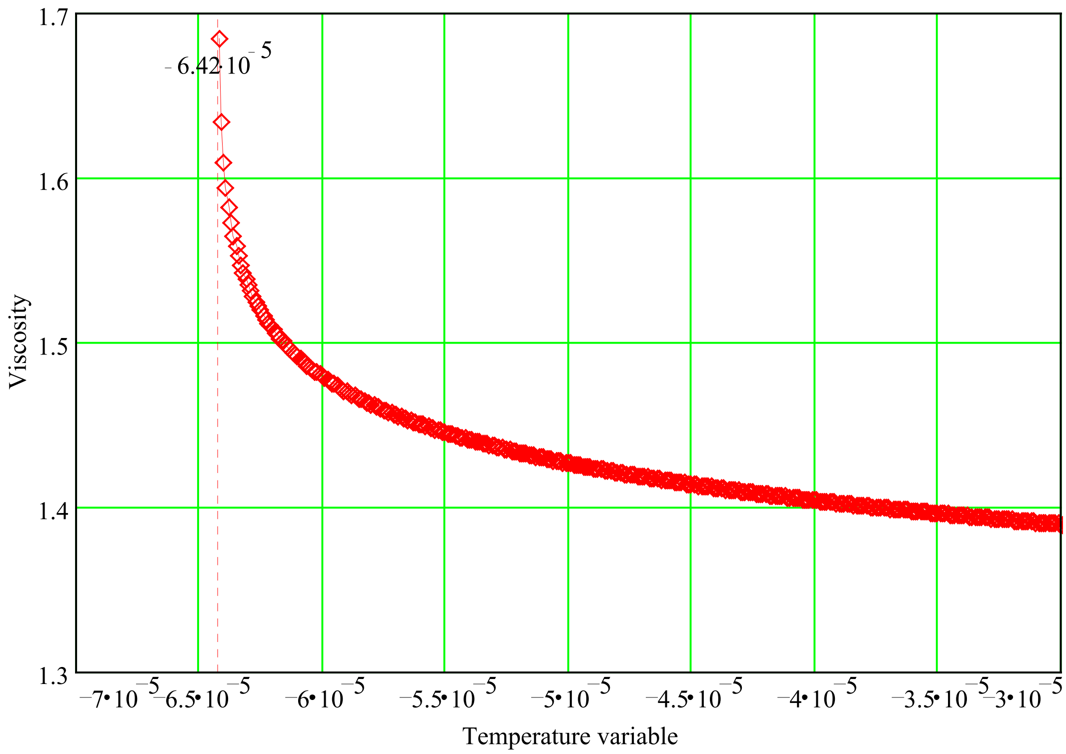

Fig. 1. Temperature dependence of the dimensionless viscosity $\eta^{*}(K)/(\eta^{B}\cdot A)$ for the cylindrical sample. Here assumed critical exponent $\nu=0.5$, $x=0.06$ and geometrical factor $K=300$.

with zero boundary condition. Our calculation shows that anomalous growth of viscosity appeared not under bulk critical temperature $T=T_c$ ($\tau=0$), but, as it was to be expected, under the new critical temperature $T_c^*(K)$, which is the same one that had been determined from Eq. (4). This fact give us an opportunity to define new critical temperature by both ways - from maximum in correlation length or from maximum in viscosity. Results which are obtained for the viscosity allow us to study the specific features of the critical behavior of the width of the central line in spectra of the light critical opalescence in finite-size liquids.

3. PROPERTIES OF LIQUID SYSTEM WITH GEOMETRY OF INFINITE BAR

Very many details about the structure and work of the "molecular machine" in biological membranes – choline receptor now are known. Choline receptor consists of five subunits - gaps. The study of amino-acid sequences of these units has shown, that all of them have taken place in an outcome of updating of the same gene. Incorporating among themselves, these subunits derivate at centre the channel. This channel has an approximately square section with the party of quadrate 0,65 nm. It does not distinguish ions K^+ and Na^+, but does not pass anions. Length of the choline receptor in 5-6 times exceeds a thickness of a membrane which is 6-10 nm, so that fiber hardly dives from it out and inside of a cell. It gives a sense of reality for geometrical model which we will consider hereafter.

The major problem of the statistical physics approach to the finite-size effects on phase transitions is the problem to find the pair correlation function and the corresponding correlation length of order parameter fluctuations. It may be mentioned that pair correlation function does in fact permit determination of most equilibrium properties of a simple liquid. Because of this the concrete purposes of conducted study are calculation and analyses of its behaviors for systems with scalar order parameters like classical liquids in the sample of rectangular section ($x\in[-a;a]$, $y\in[-b;b]$ and $z\in[0;\infty[$). The task of searching pair correlation function G_2 in this case is reduced to the finding nonsingular solution of simple boundary problem for the inversed Helmholtz operator $\hat{L}=\Delta-k^2$ associated with the Ornstein-Zernike equation [22] (Δ - is Laplasian and $k=R_c^{-1}$ is the parameter bounded to the correlation length of the infinite system), which is written for rectangular coordinates. The judicious choice of a boundary condition is important. Let is find the pair correlation function for a sample with rectangular geometry in case of a boundary condition of an constant kind, given on a surface of the bar. We shall write down a boundary condition in the form $G_2|_{x=\pm a}=A$, $G_2|_{y=\pm b}=B$ and $G_2|_{z\to\infty}=0$. Using the standard method of division of variables and omitting insignificant details of the corresponding calculations, the following solution of the given boundary problem for the general case of rectangular section of the infinite bar is received:

$$G_2(x,y,z)=\sum_{n,m=1}^{\infty}[(-1)^n(-1)^m\cdot AB\cdot Cos(\pi n x/a)\cdot Cos(\pi m y/b)]\times$$

$$\times[(1/z)\cdot exp\{-z\cdot\sqrt{k^2+\pi^2\cdot(m^2a^2+n^2b^2)/a^2b^2}\}] \qquad (16)$$

Those terms of the sum, which dumping slower than others, will define the value of the correlation length. So they will give the main contribution in the G_2. It is a reason to take into account only first member of the row with m,n=1.

For the particular case of the square section of the bar ($x,y\in[-a;a]$) with boundary condition $G_2|_{x,y=\pm a}=A$ and $G_2|_{z\to\infty}=0$ we have the main term of G_2:

$$G_2(x,y,z)=A^2\cdot Cos(\pi x/a)\cdot Cos(\pi y/a)\cdot(1/z)\cdot exp\{-z\cdot\sqrt{k^2+2\pi^2/a^2}\} \qquad (17)$$

The solution for zero boundary condition $G_2|_{x,y=\pm a}=0$ is

$$G_2(x,y,z)=\sum_{n=1}^{\infty}[D_n\cdot Cos(\pi n x/a+\pi/2)\cdot Cos(\pi n y/a+\pi/2)]\cdot[(1/z)\cdot exp\{-z\cdot\sqrt{k^2+2\pi^2 n^2/a^2}\}] \quad (18)$$

The pair correlation function G_2 varies with an exponential decay with respect to z an thus following the same way like for case with cylinder, we will defined a z-component of correlation length $(R_c)_z$ from the criteria $G_2(z)=\boldsymbol{e}\cdot G_2(z+(R_c)_z)$ and get

$$(R_c)_z=a\cdot(1+ln0.5)/(a^2k^2+2\pi^2)^{1/2} \qquad \text{for } \tau>0, \qquad \text{and}$$

$$(R_c)_z=a\cdot(1+ln0.5)/(2\pi^2-a^2k^2)^{1/2} \qquad \text{for } \tau<0 \qquad (19)$$

For such system, as well as for cylinder, the shift of critical parameters will take place. The new critical temperature $T_c^*(a)$ will depend on size a and could be defined from the condition of anomalous growth behavior of $(R_c)_z$. Thus we found

$$T_c^*(a)=T_c\cdot[1+(\sqrt{2}\cdot\pi/k_o a)^{1/\nu}]^{-1} \qquad (20)$$

Here $k_o=R_{co}^{-1}$ - the inverted amplitude of correlation length and $k=k_o\tau^\nu$. It had better to set the geometrical factor $K=k_o\cdot a=a/R_{co}$, which will characterize how mach times the cross-section's size bigger then R_{co}. Also it is possible to consider the K as number of molecular layers. In order to simplify our formulae we rewrite them using K:

$$(R_c(K))_z= R_{co}K(1+ln0.5)/(2\pi^2-K^2\tau^{2\nu})^{1/2} \qquad (21) \qquad \text{and}$$

$$T_c^*(K)=T_c\cdot[1+(\sqrt{2}\cdot\pi/K)^{1/\nu}]^{-1} \qquad (22)$$

All limit transitions to the infinite system (a, $K\to\infty$) are satisfied. On Fig. 2 the dependence of correlation length $(R_c(K))_z$ for finite-size bar system and $R_c(\infty)$ for bulk system on temperature variable τ is presented at K=100. The Fig. 3 is for dependence of the new critical temperature $T_c^*(K)$ on geometrical factor K. Here assumed critical temperature for bulk system T_c=300°K and the critical exponent ν=0.5 .

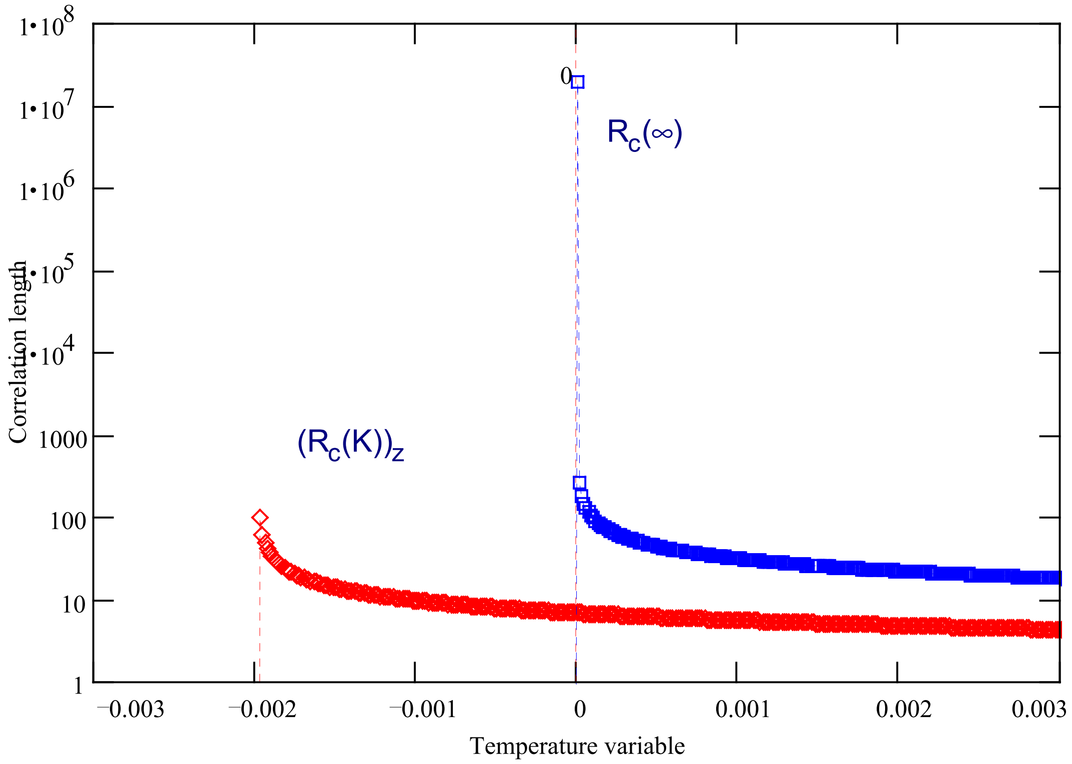

Fig. 2. Dependence of the dimensionless correlation length $(R_c(K))_z/R_{c0}$ for finite-size bar system and $R_c(\infty)/R_{c0}$ for bulk system on temperature variable $\tau=(T-T_c)/T_c$ at geometrical factor K=100.

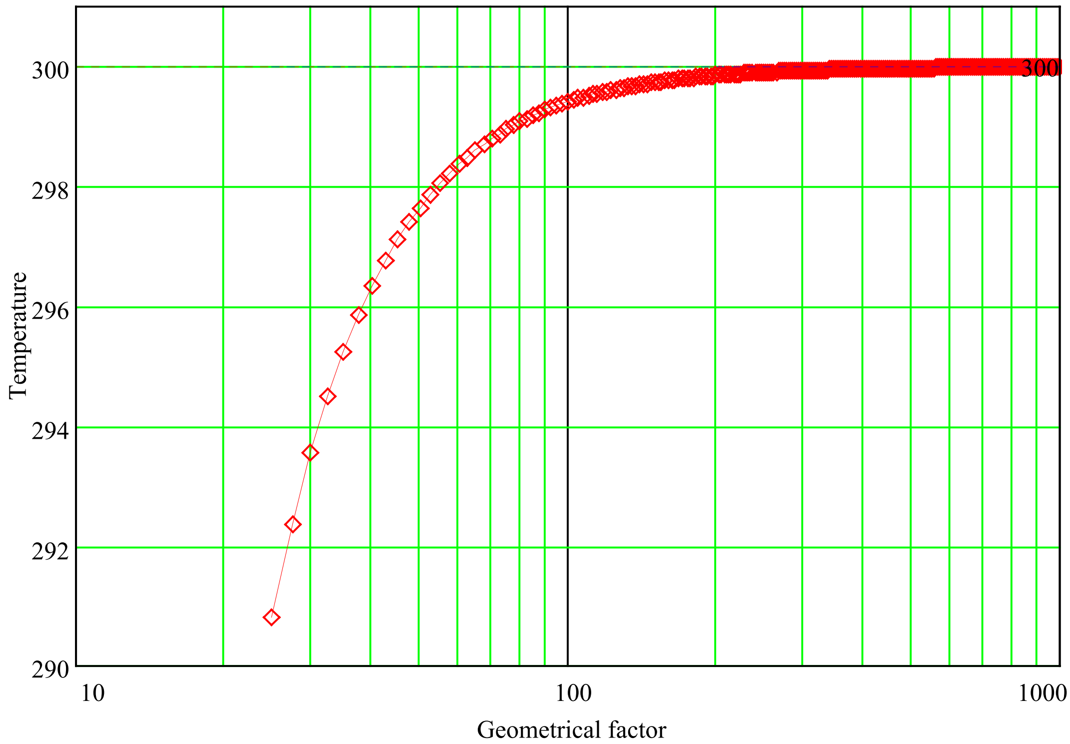

Fig. 3. Dependence of the new critical temperature $T_c^*(K)$ on geometrical factor K. Here assumed critical temperature for bulk system T_c=300°K and the critical exponent ν=0.5 .

Previous results let us to make the conclusion that reducing of the volume's size leads after the decreasing of the correlation length to decreasing of the susceptibility χ which is equivalent of isothermal compressibility β_T for one-component liquid. It could be explained in terms of well known scaling formula $\beta_T \sim R_c^{2-\varphi}$ where $\varphi=0$ for Ornstein-Zernike approximation. For obvious reason of system's spatial anisotropy we will take into account only longitudinal z-component of correlation length which highly exceed the value of component in the plane of bar section. Dependence of β_T on temperature variable τ and geometrical factor K could be describe by the equation:

$$\beta_T \sim (\tau^{\gamma}+2\pi^2/K^2)^{-1} \qquad \text{for } \tau>0, \qquad \text{and}$$

$$\beta_T \sim (2\pi^2/K^2-\tau^{\gamma})^{-1} \qquad \text{for } \tau<0 \tag{23}$$

and presented on Fig. 4. Here γ is the critical exponent. The anomalous growth of β_T manifests at the new critical temperature which is the same one, as it was to be expected, which is calculated for the present geometrical conditions of spatial limitation of a system.

4. CONCLUSION

This method and results we have presented here are general and its quantities are approximate. Of course current estimates of critical parameters, exponents far from quality of highest resolution numerical methods, but it gave us right direction of shifts and here enough room to make these results more precise by using more realistic approximation and by taking into account more factors which may play a significant role for critical phenomena in the finite-size systems.

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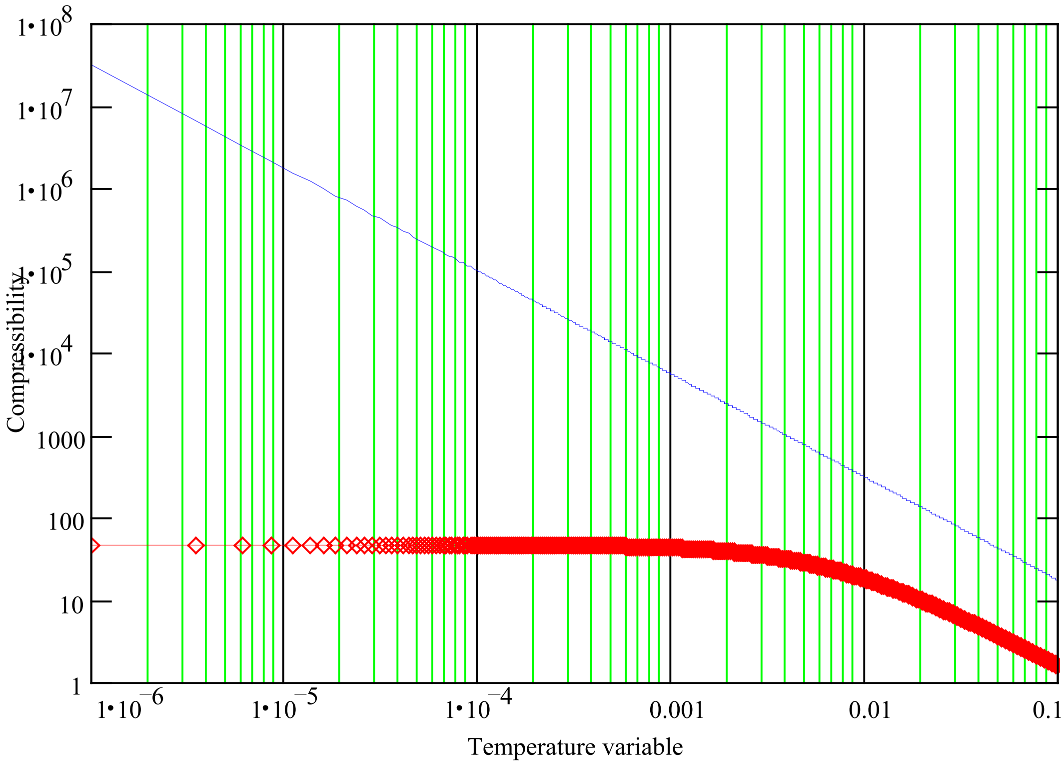

Fig. 4. Dependence of the dimensionless isothermal compressibility β_T for finite-size bar system on temperature variable $\tau=(T-T_c)/T_c$ at geometrical factor K=100. Straight line represents temperature dependence of isothermal compressibility for bulk system. Here assumed the critical exponent $\gamma=1.25$.

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